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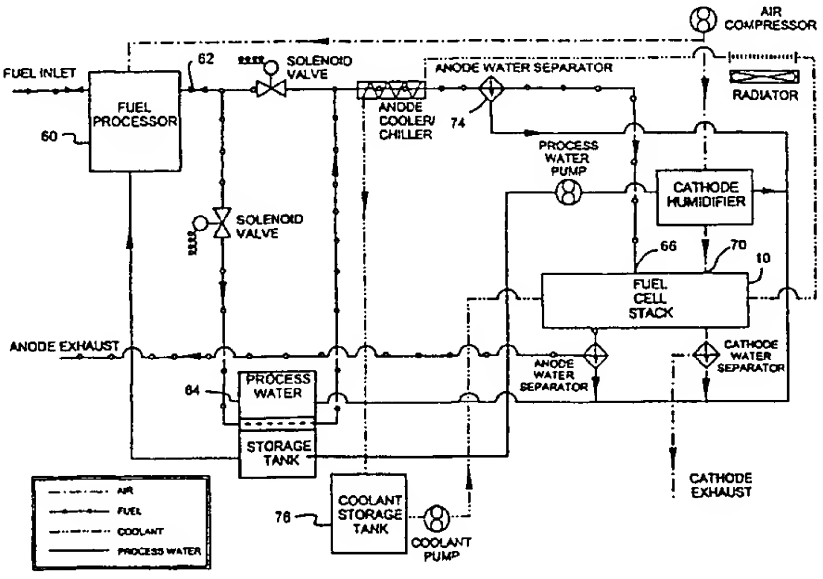

(57) Abstract

A freeze tolerant fuel cell system and method of operating the freeze tolerant fuel cell system is disclosed. The freeze tolerant fuel cell system is realized by separating the coolant loop [25] from the active membrane [12] through the use of gaskets [20, 21] interposed between the collector cell plates [18, 19]. A method of operating the freeze tolerant fuel cell system is disclosed which comprises flowing a coolant fluid other than pure water having a sufficiently low freezing point through the coolant loop [25]. A method for startup and shutdown of the freeze tolerant fuel cell system is also disclosed.

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FREEZE TOLERANT FUEL CELL SYSTEM AND METHOD

CROSS REFERENCE TO RELATED APPLICATIONS

This application is related to and claims the benefit from U.S. Provisional Application 60/130,801 entitled "FUEL CELL AND SYSTEM WITH FREEZE TOLERANT COOLANTS" filed April 23, 1999, the entirety of which is incorporated herein by reference.

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to fuel cell systems and more particularly to cooling systems for fuel cell systems.

2. Description of the Relevant Art

A fuel cell is a device that generates electrical energy by converting chemical energy directly into electrical energy by an electrochemical reaction of fuel and oxygen supplied to the cell. A typical fuel cell includes a casing which houses an anode, a cathode and an electrolyte membrane. The electrolyte membrane is disposed between the anode and cathode. A catalyst layer is disposed on the electrolyte-facing surface of each electrode. Suitable catalysts include nickel, silver, platinum and, in the case of the stabilized zirconium oxide electrolyte, base metal oxides. Platinum is most commonly used. Appropriate fuel material and oxidant are supplied respectively to the anodes and cathodes, the fuel and oxidant electrochemically react to generate an electric current, and the reaction end product is withdrawn from the cell. A relatively simple type of fuel cell (commonly called a

PEM fuel cell) uses hydrogen and oxygen as the fuel and oxidant materials, respectively. Hydrogen combines with oxygen to form water while at the same time generating an electrical current and heat. More specifically, hydrogen is consumed at the fuel cell anode releasing protons and electrons as shown in equation (1) below.

(1) $H_2 \rightarrow 2H^+ + 2e^-$ Anode Reaction

Protons produced are drawn into the fuel cell electrolyte. The electrons produced travel from the fuel cell anode to the anode terminal, through an electrical load, back to the cathode terminal, and into the cathode of the cell. A flow of ions through the electrolyte completes the circuit. Chemical reaction rates vary with location on the electrode and are dependent upon such local factors as reactant and product concentrations and temperature. At the cathode oxygen, along with electrons from the load and protons from the electrolyte combine to form water as shown in equation (2) below.

(2) $\frac{1}{2} O_2 + 2H^+ + 2e^- \rightarrow H_2O$ Cathode Reaction

The main advantage of a fuel cell is that it converts chemical energy directly to electrical energy without the necessity of undergoing any intermediate steps, for example, combustion of a hydrocarbon or carbon based fuel as in a conventional thermal power station. Fuel cells can be classified into several types according to the electrolyte used. Relatively high performance fuel cells use electrolytes such as aqueous potassium hydroxide, concentrated phosphoric acid, fused alkali carbonate and stabilized zirconium oxide.

Since individual fuel cells may produce less than the desired voltage for a

given application at full load, practical fuel cells stack several individual fuel cells in series to attain the desired voltage level by electrically connecting the cathode of one cell to the anode of an adjacent cell. Consequently, fuel cell stacks are commonly used.

The overall reaction in the cell (i.e. formation of water) is highly exothermic. The rate of heat generation is dependent upon the reaction rate and the heat flux across a given area of the fuel cell and is proportional to the local reaction rate. Consequently, a structure for cooling a fuel cell is generally required and is generally designed based on the projected peak heat flux. Water is generally used for cooling fuel cells.

PEM fuel cells generally require humidification to maintain the moisture of the electrolyte membrane which is required for efficient fuel cell operation. A single water loop generally provides both humidification and cooling for fuel cells.

In some fuel cell applications, such as automotive applications, it may be necessary to commence operation of a fuel cell stack having a core temperature below the freezing temperature of water. For example, the SAE automotive standard requires operation between -40°C and 53°C and survival (storage) of between -46°C to 66°C. Numerous difficulties are encountered in attempting to operate a fuel cell in applications below the freezing point of water. Water is known to expand significantly upon freezing. Ice formation inside the fuel cell system may destroy fuel cell components. Even if component damage does not occur upon freezing, blockage of fuel system lines may occur which can delay the startup of the fuel cell. Finally, for sub-freezing applications, coolants must be selected that freeze

at temperatures below the freezing point of water. Most available coolants which have freezing points below the freezing point of water are known to "poison" the catalyst layer by binding to catalyst sites if such materials are allowed to come in contact with the catalyst layer. What is needed is a freeze tolerant fuel cell design and methodology that permits reliable fuel cell operation at temperatures well below the freezing temperature of water.

SUMMARY OF THE INVENTION

It is accordingly an object of the invention to provide a fuel cell system that can be used in sub-freezing environments.

It is another object of the invention to provide a fuel cell system that can utilize coolants other than a coolant compatible with the MEA.

It is yet another object of the invention to provide techniques for removing water from the fuel cell on shut down and rapidly reheating the fuel cell upon start up.

These and other objects of the invention are achieved by a number of embodiments incorporating features and advantages of the invention. The invention can include a freeze tolerant fuel cell system including at least one fuel cell made up of a pair of collector plates having a series of channels for the flow of reactants. A first and a second gas diffusion layer is disposed between said collector plates; and a membrane electrode assembly (MEA) including a membrane sandwiched between two electrode layers. The MEA is interposed between said gas diffusion layers, and the reactant's on each side of the MEA are substantially sealed from leaking to the

other side of the MEA. The fuel cell stack can further include at least one coolant passage for flowing a coolant stream relative to the collector plates so that the coolant stream does not contact said MEA while cooling the fuel cell.

According to another aspect of the invention, the coolant flowing in said coolant passage is poisonous to the MEA and thus must be mechanically isolated from the MEA. The surfaces of the coolant passage in contact with the coolant can be electrically insulated or the coolant can be electrically non-conductive. The isolation can be provided by a gasket arrangement around coolant ports running through the collector plates. The gaskets are preferably spaced from the gas diffusion layers. Alternatively, the cooling passage can be positioned outside the conductive portion of the collector plates and surrounded by housing. This housing can be molded integrally with the collector plates or otherwise connected to the collector plates.

The fuel cell coolant loop can include edge cooling within the fuel cell, or a coolant layer outside said collector plates can be provided. This coolant layer can be directly adjacent each active fuel cell or provided at intermittent stages throughout a fuel cell stack.

According to another aspect of the invention, the interface region between the peripheral port gaskets and the active area membrane can be supported by bridging sub-gaskets to avoid damaging mechanical and electrical edge effects in the membrane. In preferred embodiments, the sub gaskets extend across the uncovered membrane region between the port gaskets and the gas diffusion layers. The sub-gaskets can be made of a number of materials, including FEP, TFE, ETFE,

PFA, CTFE, E-CTFE, PVF_2 and PVF.

To further equip a fuel cell system for use in sub-freezing environments, the invention further contemplates improvements and techniques relating to shut down and start up to avoid the presence of significant quantities of water in the fuel cell when the fuel cell temperature falls below the freezing point of water, such as when the fuel cell system is not operating.

During fuel cell operation, water can accumulate in the gas diffusion layers as well as the flow channels of the collector plate. According to one aspect of the invention, the reactant channels in the collector plates are discontinuous, whereby flow fields are established through the gas diffusion layers. With this arrangement, as part of a freeze tolerant shut down operation, purging dry gases can be forced through the fuel cell, collecting and driving water not only out of the channels but also the gas diffusion layers.

To further reduce the accumulation of residual water after shut down, the surfaces of the channels are preferably essentially impermeable to water. Further, the surfaces of the channels can be essentially impermeable to all fluids.

To further assist in the effective removal of water during shut-down, a fuel cell system according to the invention can provide counterflow in the gas diffusion layers on either side of the membrane to increase the moisture gradient and rate of water transfer. The reactant channels of each collector plate can be arranged so the direction of reactant flow in one gas diffusion layer is opposite the direction of reactant flow in the other gas diffusion layer.

Another improvement to assist in water purging relates to the positioning of

the outlet channels to use gravitational force. A fuel cell system according to the invention can have reactant outlets in which at least one of the outlets is positioned below the channels, whereby water removal is assisted by gravitational force.

The drained water can be removed from the system or collected in a reservoir, such as a tank. The tank can be rendered freeze tolerant in a number of ways, including insulation or heating from either a direct, dedicated source, or a temporary source, such as heat production from the system's fuel processor or the fuel cell. During dormant steps, watering in the tank can be allowed to freeze, provided the tank is designed to permit expansion of freezing water.

To protect against the mechanical damage to the collector plates due to the freezing expansion of any residual water, the walls of the channels can be tapered and have rounded corners.

According to the invention, a fuel cell system can be made more freeze tolerant by incorporating shut down procedures conducive to freezing environments. The shut-down procedure can include the steps of: reducing the fuel cell system temperature, whereby water vapor in said fuel stack is condensed; removing water, liquid and gaseous, from said fuel cell; purging said the reactant gas lines with a non-humidified gas; and reducing the system pressure to a pressure approximately equal to atmospheric pressure. These steps can be performed in different orders, and alternatively, can be begun simultaneously.

The shut down procedure can also include steps to further increase heat of the fuel cell at or just before shut-down. One preferred step includes running said fuel stack in a mode that results in a pulsed current output.

The presence of substantial quantities of water during start-up, before the fuel has heated to above the freezing point, can present a danger of damage by freezing expansion of the water. So, techniques according to the invention also provide for start-up procedures that introduce water so as to avoid freezing. These techniques can include the steps of: flowing dry reaction gases through the reactant lines into said fuel cell; measuring the temperature of said fuel cell; and initiating humidification of said reactant gases after the fuel cell temperature is above a predetermined temperature. The predetermined temperature can be the freezing point of water at the ambient pressure. The start-up procedure can also preferably include pressurizing to maximum operating pressure to increase heating of the fuel cell and retain any thawing residual water.

The start-up procedures can also include: providing a humidifier for humidifying reactant gas flows to the fuel cell; providing a water reservoir for the supply of water for humidification of the reactant gas flows; heating the reservoir to melt the water in the reservoir; melting water in the reservoir; and supplying water to the humidifier for humidification of the gas flows.

The heat for melting the water in the reservoir can be obtained from the fuel processor. The steps of an implementing method can include: operating a fuel processor in an oxidant rich mode to increase heat output; transferring a portion of said heat output to said reservoir to melt water in the reservoir; transferring a portion of said heat output to said fuel cell to raise the fuel cell temperature; monitoring the melting of the water in said reservoir and the fuel cell temperature; adjusting the reactant mixture in the fuel processor to increase fuel production after at least a

portion of the water in said reservoir is melted and the fuel cell temperature has reached a predetermined temperature.

All the above constructions and operating methods can be employed in various combinations. Alone and in combination, these features contribute to the utilization of a fuel cell system in sub-freezing environments.

BRIEF DESCRIPTION OF THE DRAWINGS

Features and advantages of the present invention will become apparent to those skilled in the art from the following description with reference to the drawings, in which:

Fig. 1 illustrates a breakaway side view of fuel cell and coolant system according to the invention.

Fig. 2 illustrates a freeze tolerant fuel cell system schematic comprising a fuel cell stack interfaced with a fuel processor and external cooling and humidification systems used to implement Applicants' method of using a freeze tolerant fuel cell.

Fig. 3 illustrates a side view of a fuel cell having collector plates with non-continuous flow field channels with tapered and rounded corners.

Fig. 4 illustrates a breakaway side view of a fuel cell having primary gaskets and secondary sub-gaskets.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

A novel freeze tolerant fuel cell structure is provided that is adapted for operation in environments subject to sub-freezing temperatures, hereinafter referred to as sub-freezing environments. As used throughout this specification, sub-freezing environments refers also refers to ambient conditions that may be above freezing during operation of a fuel cell, but reach sub-freezing temperatures are uring dormant periods of the fuel cell. The cooling system and humidification systems are preferably separated. The cooling system is also preferably isolated from the electrochemically active region of the fuel cell through the use of gaskets. By separating the cooling and humidification systems and isolating the cooling system from the electrochemically active fuel cell region, materials otherwise poisonous to the fuel cell which have freezing points below the freezing point of water may be used as coolant materials. A method for operating the freeze tolerant fuel cell during sub-freezing conditions is also disclosed. During the startup of the fuel cell from an idle mode during sub-freezing conditions, the fuel cell system is run in a mode that maximizes the heat generated by the fuel cell. Humidification is delayed until the fuel cell temperature is raised above the freezing temperature. Shutdown of the fuel cell during sub-freezing conditions involves removing as much water as possible from the fuel cell in a minimum period of time.

A novel freeze tolerant fuel cell structure having both gaskets and sub-gaskets is also disclosed. Upon assembly of the freeze tolerant fuel cell, an interface region between the gaskets and the edge of the electrochemically active region of the fuel cell is formed. This region is subject to enhanced mechanical and enhanced

electrical stress due to increased edge conduction relative to electrochemically active areas of the fuel cell removed from the interface region. Use of sub-gaskets in conjunction with gaskets in the fuel cell is disclosed which minimizes the mechanical and electrical stress at the edges of the electrochemically active fuel cell and leads to improved fuel cell reliability.

Referring to FIG. 1, a fuel cell is identified generally by the reference numeral 10. A single fuel cell 10 depicted in the drawing, comprised of one cell unit 11. However, it is to be understood that the invention can be utilized in conjunction with fuel stacks having a plurality of cell units. Each cell unit 11 includes a membrane electrode assembly (MEA) 12 comprised of a solid ion conducting membrane which may have an anode 13 on one side and a cathode 14 on the other side of the membrane, each formed by an electrochemically active catalyst layer attached directly to the outside of the membrane. The MEA 12 is interposed between a first gas diffusion layer 15 and a second gas diffusion layer 16.

Alternatively, the anode and cathode may be attached or integrated into the gas diffusion layers 15 and 16, and pressed against the membrane. In the preferred embodiments, the MEA includes an attached anode 13 and cathode 14 due to greater fuel cell efficiency resulting from intimate contact of the electrodes with the membrane compared to a pressure contact when the electrodes are mounted on the gas diffusion layers 15 and 16. Preferably, the MEA 12 extends some minimum distance beyond the outer periphery of the gas diffusion layers 15 and 16. Alternatively, the MEA can terminate with the edges of the gas diffusion layers, provided that edge is properly sealed to prevent the leakage of reactants around the

MEA. The gas diffusion layers 15 and 16 are interposed between two electrically conducting collector/separator plates 18 and 19 (collector plates), which are commonly referred to as bipolar plates when two or more fuel cells are used to form a fuel cell stack. The gas diffusion layers 15 and 16 are typically fabricated from porous, electrically conductive materials, such as carbon/graphite fiber paper or carbon/graphite cloth. When two or more fuel cells form a fuel cell stack, the collector plates 18 and 19 are provided for separating the cathode of one cell unit 11 from the anode of adjoining cell units (not shown).

In the preferred embodiment of the invention, the fuel cell 10 is a proton exchange membrane (PEM) fuel cell. Since individual PEM fuel cells produce less than 1 volt at full load, practical PEM fuel cells stack several individual cells, such as cell unit 11 in series to attain the desired voltage level by electrically connecting the cathode of one cell to the anode of an adjacent cell (not shown).

The collector plates 18 and 19 are electrically conductive. In the preferred embodiment of the invention, the collector plates 18 and 19 are formed from electrically conductive polymer composites by filling a polymer with a plurality of conductive particles, such as graphite. The collector plates 18 and 19 may be designed to have a spectrum of water permeabilities from highly permeable to essentially impermeable. In some applications, it may be desirable to have water permeable collector plates. For example, water permeable collector plates may provide water vapor to the fuel cell for membrane humidification. However, during fuel cell operation, water permeable collector plates store significant quantities of water. In fuel cell applications at temperatures below the freezing point of water,

water impermeable collector plates 18 and 19 are generally required to avoid damaging the collector plates 18 and 19 through expansive forces exerted by freezing water contained therein. Accordingly, in the preferred embodiment of the invention, collector plates 18 and 19 selected are essentially impermeable to water. At a minimum, the surfaces of the channels are impermeable to water while the remainder of the collector plate may be permeable to water.

The electrochemical reaction at the cathode 14 that forms water is highly exothermic. Consequently, a cooling system is generally required for fuel cells. De-ionized water is commonly used to cool fuel cells and also to maintain the hydration of the membrane ("humidifying"), which is known in the art to be required for efficient ionic transport across the membrane. Although this arrangement is quite satisfactory in most fuel cell applications, in applications where the temperature may reach freezing temperatures (at or below 0°C at one atmosphere pressure), pure water cannot be used as a coolant because if water freezes the fuel cell will be damaged or destroyed by the associated expansion of water during its phase change to solid form. Membrane humidification, if required, must also be redesigned to avoid freezing water in the fuel cell 11. If humidification is required, the invention separates the humidification and the coolant systems. Separation of humidification and cooling permits use of coolants other than pure water. This is important, since the coolant fluid in a freeze tolerant fuel cell cannot be pure water since water freezes at 0°C at 1 atmosphere pressure.

Humidification of the membrane may be derived from a source outside the fuel cell stack, such as by humidifying incoming reactant gases through the use of

misters or bubblers. In situations where fuel processors are used to produce hydrogen fuel, the anode may not require humidification, due to moisture produced as a by-product of the reforming process.

A dedicated coolant loop 25 has a coolant flow field through and between collector plates, which form a cooling layer adjacent to the fuel cell 11. Electrically conductive sealant 32 binds top collector plate and bottom collector plate. Coolant flows both vertically though the passage and laterally through the cooling layer to dissipate heat uniformly across the collector plate surface area of the adjacent fuel cell 11. A coolant return path is provided but not shown.

Coolant loop 25 does not provide humidification to the fuel cell 11. Additionally, the coolant loop 25 is isolated from the membrane by a minimum distance "A." Depending on the anticipated operating temperature range the fuel cell 11 will be subjected to, coolants other than pure water are needed as coolants for the fuel cell 11. Since water freezes near 0°C at one atmosphere pressure, pure water is not a viable coolant for sub 0°C fuel cell applications. The coolant system is mechanically sealed during manufacture so that the coolant fluid is isolated from the membrane. The distance "A" is chosen to avoid coolant contact with the membrane and is based on the integrity of the overall fuel cell seal. In the preferred embodiment of the invention, the distance "A" is at least approximately 0.1 inches. Seal integrity is principally a function of the type of gasket material selected.

Many coolants other than pure water will contaminate the fuel cell if allowed to contact the membrane by taking up catalyst sites. For example, hydrocarbons are known to occupy catalyst sites. As used throughout the specification, these

contaminating coolants are referred to as "poisonous." Poisonous coolants are intended to refer to all non-pure water coolants that can bind the catalyst if allowed to contact the MEA electrodes or otherwise interfere with the electro-chemical reaction of the fuel cell.

In an alternate embodiment of the freeze tolerant fuel cell, the coolant passage way is not part of the collector plate. For example, coolant may be flowed through an external manifold in proximity to the periphery of the fuel cell collector plates. In yet an another embodiment of the fuel cell, coolant channels are placed on the periphery of collector plates in areas removed from the fuel cell active area. In both of these alternate embodiments, coolant does not pass between the region between adjacent collector plates.

Coolants other than pure water having a freezing point sufficiently below 0°C to meet the expected minimum temperature of operation of the fuel cell 11 will be generally useful for freeze tolerant fuel cells. Possible suitable coolants include:

(a) ethylene glycol alone, with any percentage of other coolants including water, and/or lubricants, provided the freezing point of the solution is sufficiently low for its intended application.

(b) propylene glycol alone, with any percentage of other coolants including water, and/or lubricants, provided the freezing point of the solution is sufficiently low for its intended application.

(c) other alcohols with similar freezing points and boiling points to propylene glycol and ethylene glycol such as methanol, with any percentage of other coolants including water, and/or lubricants, provided the freezing point of the solution is

sufficiently low for its intended application; and

(d) gases under anticipated conditions of operation, such as nitrogen or hydrogen.

The maximum allowable coolant ionization level depends on the design of the coolant loop 25. If the coolant loop 25 is designed to be electrically isolated from the collector plates 18 and 19, ionic coolants may be used. However, if the coolant loop 25 is not designed to be electrically isolated from the collector plates 18 and 19, the coolant ionization level must be limited to avoid electrically coupling neighboring collector plates 18 and 19 (not shown) through the fluid flowing through the coolant loop 25. If neighboring collector plates 18 and 19 (not shown) are at different electrical potentials and are electrically coupled through a coolant loop 25 that uses a conductive coolant material, current will flow between neighboring collector plates 18 and 19 (not shown) through the conductive coolant. In the preferred embodiment of the invention, the coolant loop 25 is not electrically isolated from collector plates 18 and 19 because coolant plate and flow field design is much more complex and expensive if the coolant loop 25 is designed to be electrically isolated from collector plates 18 and 19.

The coolant isolation can be provided by a gasket arrangement. The outer edge of the MEA 12 is interposed between a first gasket 20 and a second gasket 21. Gaskets 20 and 21 are preferably positioned so as to not overlap with gas diffusion layers 15 and 16. Gaskets 20 and 21 may be made from polymer materials such as EPDM rubber (also known as EP rubber), fluorinated hydrocarbon, butyl rubber, fluorosilicone, polysiloxane, thermoplastic elastomers such as blends containing

polypropylene and EP rubber, and or other similar materials. An interface region 22 is formed between the gas diffusion layers 15 and 16 and the gaskets 20 and 21.

The membrane at or near the interface region 22 is subjected to both enhanced mechanical and electrical stress relative to other portions of the active membrane. If a space between the gas diffusion layers 15 and 16 and the gaskets 20 and 21 results during the manufacture, of the fuel cell 11, e.g. due to tolerances, the membrane in the interface region 22 will be unsupported and will tend to sag or pinch. Sagging or pinching may produce the undesirable result of a rupture which could allow reactants to flow from one flow field into another flow field. The magnitude of the mechanical stress at the interface region 22 may be reduced by effectively splitting the interface region 22 into two regions. The interface region 22 may be effectively split by shifting the upper interface between the gas diffusion layer 15 and gasket 20 and the lower interface between gas diffusion layer 16 and gasket 21 during the manufacture of the fuel cell 11. Also, during manufacture, the gas diffusion layers 15 and 16 may butted up against the gaskets 20 and 21 and result in compression of the membrane in the interface region 22. In either case, the membrane is subjected to additional mechanical stress in the interface region and may result in early life failures of the fuel cell 11.

Further, since the edges of parallel conducting plates are known to generate higher electrical field fluxes compared to interior portions of conducting plates, the active membrane at or near the interface region 22 will experience higher current densities in fuel cell 11 operation compared to interior portions. This phenomenon enhances electrical stress to the membrane at the interface region, regardless of

whether a space is formed as in Fig. 1 or the gas diffusion layers 15 and 16 butted up against gaskets 20 and 21.

Even a well designed freeze tolerant fuel cell requires special startup, shutdown and humidification processes to minimize damage resulting from the formation of ice from water trapped therein during freezing conditions. In the preferred embodiment of the invention, all operational aspects of the freeze tolerant fuel cell system, such as reactant flow control, temperature monitoring and control are controlled by a central computer using feedback control systems. Now referring to Fig. 2, to begin startup of the fuel cell stack 10, heat from the fuel stream can be first supplied to the fuel cell 10 by fuel processor 60 through anode inlet 62 with minimum water content during startup to minimize the amount of ice formed. For example, a partial oxidation based auto-thermal fuel processor propane into a hot gas stream. The percentage of hydrogen in the reformate stream may be adjusted to the desired level by varying the steam to carbon ratio as well as the stochiometric ratio of air to fuel in the fuel processor 60. High water yield is accomplished by feeding air to the fuel processor 60 at quantities close to or preferably exceeding quantities required for complete combustion of the fuel supplied. The hot gas stream produced by the fuel processor 60 is then cooled by the coolant in the anode 66, resulting in condensation of water which may be separated in the anode separator 68 and directed to the process water tank 64. Combustion is usually a very quick process and the fuel processor 60 is operated in this mode until sufficient water is collected in the process water tank 64. Process water stored in the water tank 64 will be used for steam reforming and cathode gas humidification when the

operating temperatures in the system rise above freezing point of water. In start up situations where the process water tank 64 initially contains ice, the fuel processor 60 will be operated in a combustion mode to produce maximum water as described earlier and the hot gas stream produced by the fuel processor 60 can be used to melt the ice in the freeze resistant process water tank 64. By following either of the above startup procedures the fuel processor 60 will generate a hydrogen-rich hot reactant stream to the fuel cell stack 10 with minimum moisture content during startup. A hot reactant stream will also help thaw out the various fuel stack components.

Humidification should be delayed until the stack and coolant temperatures are above the freezing point of water. During the initial stages of startup, pressurized dry air is fed to the cathode 70 without humidification. Pressurized dry air from the cathode compressor is typically heated to a temperature in the range of 90-100°C by pressurizing at 30 psig when the ambient air temperature is in the range of approximately -10 to -20°C. The hot cathode air will also help thaw out the membrane and other stack components that are exposed to freezing conditions. Once hydrogen from fuel processor 60 is available for power generation the fuel cell stack 10 will be operated in a low voltage/high current density mode to maximize generation of heat. Heat generated will be used to raise the temperature of the stack 10 and the coolant. As the stack 10 temperature increases, the stack will be operated to begin producing higher output voltages. Thus, the system may be transitioned to its more efficient electrical power generation mode as the fuel cell stack 10 temperature increases. Cathode air humidification may be begun after the

stack 10 and coolant temperature are well above freezing. Optionally, anode humidification may be begun at the same time.

In an alternate embodiment of the freeze tolerant fuel cell system, the fuel processor shown in Fig. 6 is replaced by an essentially pure hydrogen source. Hydrogen is supplied to the anode of the fuel cell along with an oxygen source to the cathode of the fuel cell to immediately generated heat. The heat generated raises the temperature of the freeze tolerant coolant. If the water tank contains ice, the heated freeze tolerant coolant is circulated through the water tank to melt the ice in the water tank. Once the stack temperature rises above the freezing point of water, humidification of the reactant lines may be begun.

A method for shutting down the freeze tolerant fuel cell is also required to avoid formation of ice within the fuel cell system upon shutdown. In the event of a system shut down, the main objective is to remove most water from within the fuel cell stack and system components as quickly as possible. There are three steps required to remove the water from the system. First, the system temperature must be reduced to condense water vapor within the system. The temperature may be reduced more rapidly by flowing a coolant through the coolant loop. Cooling the system allows water contained in vapor phase fuel cell to condense. Once water is in a liquid phase it can easily be separated from the gas streams and drained into a freeze resistant storage tank 64. Second, the fuel cell stack 10 and system reactant lines must be thoroughly purged with non-humidified gases using either an on-board or supplemental purging system. Finally, the whole system must be returned to ambient pressure. Following this three step procedure will significantly reduce the

amount of condensed water that will remain in the fuel cell after shut down. If water is left in the fuel cell during shutdown, it will expand upon freezing and may cause damage to vital fuel cell components such as the anode water separator(s), cathode water separator(s), fuel cell stack, fuel processor water storage tank (when used) and reactant supply lines.

A similar method for anode shutdown is described below. If anode humidification is used, humidification of the anode gas is stopped. The anode gas lines 62 are then purged with dry cold anode gas. This will purge all humidified anode gas from the fuel cell components and will also eliminate most liquid water droplets from the fuel cell system 10 components.

Condensed water droplets will be separated in the anode separator(s) and can either be drained into freeze tolerant storage tank 64 or be completely purged from the system. If a process water storage tank is used, the process water storage tank should be placed at the lowest possible level to permit gravity feeding into the water storage tank.

A similar method for fuel processor shutdown is described below. The fuel cell stack temperature is reduced to the ambient temperature. An anode cooler/chiller is used to condense water out of the anode stream and transfer heat to the coolant. This water is eliminated from the anode stream by an anode separator 74. The separated water can either be drained into a freeze resistant storage tank 64 or be completely purged from the system. The anode cooler/chiller holds the system coolant, which is at the stack temperature, running through it. The stack temperature is brought down to ambient such that the anode gas temperature can

also be cooled to near ambient temperature.

A similar method for cathode shutdown is described below. Cathode shut-down should be initiated at the same time as the anode shut-down. Humidification of cathode gas is terminated. The cathode system is then purged with dry cathode gas. The compressor temperature and pressure are brought down to near ambient conditions. All liquid water is separated in the cathode separator(s) and can either be drained into the freeze resistant water storage tank 64 or be completely purged from the system.

A similar method for coolant loop shutdown is described below. The system layout should be such that the coolant storage tank 76 is at the lowest point in the system. This allows the coolant to drain via gravity back into the coolant storage tank 76. At the same time as the coolant is gravity fed into the coolant storage tank, the system completes the dry reactant purge. Finally, the coolant pump is switched off.

In an enhancement of the shutdown process, the fuel cell stack 10 is operated in a mode that produces current through the stack sufficient to generate large amounts of heat while supplying the fuel cell with dry reactants. This condition will maximize the evaporation rate of water within the fuel cell stack 10 and result in a faster shut down time.

The start up and shut down procedures described include a number of steps that can be performed in different orders and can also be started simultaneously.

Referring to Fig. 3, In a further enhancement to the shutdown process, a preferred embodiment of the fuel cell is shown having interdigitated discontinuous

channels in the collector plates 18 and 19. Flow field anode inlet channel 26, cathode inlet channel 28, anode outlet channel 27 and cathode outlet channel 29 are separated thus forcing flows through the gas diffusion layers. Channels shown have rounded corners. Interdigitation and rounded corners of the flow fields need not be combined to produce fuel cell improvements for freeze tolerant applications. An interdigitated flow field configuration helps limit the amount of water trapped within the gas diffusion layers 15 and 16 and speeds the purging of water out from gas diffusion layers 15 and 16 during shut down of the fuel cell during freezing conditions. In addition, the walls of the channels making up the flow field may be tapered slightly and corners rounded to allow room for water to expand upon freezing to minimize damage to the fuel cell 11 during freezing conditions.

Fig. 4 adds a pair of sub-gaskets 23 and 24 to Applicants' gasketed fuel cell as shown in Fig. 1. Sub-gaskets 23 and 24 are positioned between first and second gaskets 20 and 21 and extend into a position between the gas diffusion layers 15 and 16 and the membrane on the distal end of the region where the gas diffusion layers 15 and 16 overlap the membrane. Sub-gaskets 23 and 24 provide extra support for the fuel cell in the interface area reducing mechanical stress on the membrane at or near the interface region 22. In addition, enhanced edge conduction across the membrane at the interface 22 is eliminated by shifting the effective edge of the active membrane to the distal end of sub-gaskets 23 and 24. Thus, the use of sub-gaskets 23 and 24 results in improved fuel cell reliability. In the preferred embodiment of this invention, sub-gaskets 23 and 24 are made from strong acid resistant materials such as FEP, TFE, ETFE, PFA, CTFE, E-CTFE, PVF_2

and PVF and have a thickness of approximately 1/10 the thickness of gaskets 20 and 21.

As shown, the coolant loop 25 passes through gaskets 20 and 21 as well as sub-gaskets 23 and 24. As discussed earlier, sub-gaskets 23 and 24 reduce mechanical and electrical stress near the edge of the active membrane in the interface region 22. Sub-gaskets 23 and 24 need not extend to be co-terminus on the side opposite the active membrane with the edge of primary gaskets 20 and 21. In the preferred embodiment of the invention, sub-gaskets 23 and 24 are co-terminus on the side opposite the active membrane with the edge of primary gaskets 20 and 21 due to ease of manufacture coupled with the low relative cost of the sub-gasket 23 and 24 material as compared to the added labor cost to construct comparatively short sub-gaskets 23 and 24 that do not extent significantly outside the active membrane region.

While the preferred embodiments of the invention have been illustrated and described, it will be clear that the invention is not so limited. Numerous modifications, changes, variations, substitutions and equivalents will occur to those skilled in the art without departing from the spirit and scope of the present invention as described in the claims.

What is claimed is:

1. A freeze tolerant fuel cell system, comprising at least one fuel cell, said at least one fuel cell system comprising:

a pair of collector plates having a series of channels for the flow of reactants from ports formed through the collector plates;

a first and a second gas diffusion layer disposed between said collector plates; and

a membrane electrode assembly (MEA) including a membrane sandwiched between two electrode layers, said MEA being interposed between said gas diffusion layers, a seal being provided to substantially prevent the transfer of reactance gases around the MEA;

said fuel cell system further comprising at least one coolant passage for flowing a coolant stream relative to said collector plates to cool said fuel cell and wherein said coolant stream does not contact said MEA.

2. The fuel cell system as recited in claim 1, further comprising a coolant flowing in said coolant passage, said coolant being poisonous to the MEA.

3. The fuel cell system as recited in claim 1, wherein surfaces of the coolant passage in contact with the coolant are electrically insulated.

4. The fuel cell system as recited in claim 1, wherein the coolant is electrically non-conductive.

5. The fuel cell system as recited in claim 1, further comprising a coolant loop, said at least one coolant passage forming part of said coolant loop.

6. The fuel cell system as recited in claim 5, wherein said coolant loop includes

a coolant layer outside said collector plates.

7. The fuel cell system as recited in claim 6, wherein said coolant layer is adjacent one of said collector plates.

8. The fuel cell system as recited in claim 5, wherein the coolant loop includes coolant channels in said collector plates, separate from said reactant channels.

9. The fuel cell system as recited in claim 1, wherein said collector plates provide separate coolant ports for the transfer of coolant apart from the reactant flows and further comprising a pair of gaskets surrounding the coolant ports and disposed between the collector plates.

10. The fuel cell system as recited in claim 1, wherein said coolant passage is housed in structure outside the conductive region of said collector plates.

11. The fuel cell system as recited in claim 9, wherein the pair of gaskets is separated from the gas diffusion layers.

12. The fuel cell system as recited in claim 9, wherein at least said membrane extends beyond at least a portion of the periphery of said gas diffusion layers and the pair of gaskets overlap the membrane.

13. The fuel cell system as recited in claim 9, further comprising a pair of sub-gaskets, each sub-gasket positioned between said gaskets and extending into a position between said gas diffusion layers and said MEA, whereby said membrane is supported.

14. The fuel cell system as recited in claim 13, wherein said sub-gaskets are made from a fluoro-polymer.

15. The fuel cell system as recited in claim 14, wherein said sub-gaskets are

made materials selected from the group consisting of FEP, TFE, ETFE, PFA, CTFE, E-CTFE, PVF_2 and PVF.

16. The fuel cell system as recited in claim 1, wherein said fuel cell is a proton exchange membrane (PEM) fuel cell and said ion exchange membrane is water permeable.

17. The fuel cell system as recited in claim 1, wherein said reactant channels are discontinuous, whereby flow of reactant gases are established through the gas diffusion layers.

18. The fuel cell system as recited in claim 17, wherein said reactant channels in each collector plate are arranged so that the direction of reactant flow in one gas diffusion layer is opposite the direction of reactant flow in gas diffusion layer on the opposite side of the MEA, whereby a greater moisture gradient is established to facilitate the transfer of water during shutdown, purging operations.

19. The fuel cell system as recited in claim 17, wherein said reactant channels in each collector plate are arranged so that relatively dry portions of the reactant flow on one side of the MEA oppose relatively wet portions of the reactant flow on the other side of the MEA, whereby a greater moisture gradient is established to facilitate the transfer of water during shutdown, purging operations.

20. The fuel cell system as recited in claim 1, wherein surfaces of the channels are essentially impermeable to water.

21. The fuel cell system as recited in claim 20, wherein surfaces of the channels are essentially impermeable to all fluids.

22. The fuel cell system as recited in claim 1, wherein walls of said channels are

tapered and have rounded corners, whereby any residual water in the fuel cell that freezes can expand freely and avoid damage to the collector plates.

23. The fuel cell system as recited in claim 1, wherein the reactant channels have reactant outlets, at least one of said outlets being positioned below the channels, whereby water removal is assisted by gravitational force.

24. The fuel cell system as recited in claim 1, further comprising a water reservoir positioned outside the stack, wherein the reactant channels have reactant outlets operatively connected to the water reservoir to permit water from the fuel cell to drain to water reservoir.

25. The fuel cell system as recited in claim 24, wherein the water reservoir is freeze tolerant, including at least one of insulation and a heating source.

26. The fuel cell system as recited in claim 25, wherein the heating source is a heater for the reservoir.

27. The fuel cell system as recited in claim 25, wherein the heating source is a fuel processor.

28. The fuel cell system as recited in claim 25, wherein the heating source is the fuel cell.

29. A method of using a fuel cell system in environments subject to sub-freezing temperatures, said fuel cell system having at least one fuel cell, said fuel cell including a membrane, a pair of gas diffusion layers, a pair of electrodes, a pair of collector plates, at least two reactant flow lines, comprising the steps of:

creating at least one coolant flow passage relative to said fuel cell so as to avoid coolant contact with said membrane while cooling the fuel cell;

flowing a coolant stream poisonous to the membrane through said at least one passage, whereby said coolant stream does not contact said membrane such that a coolant having a freezing point below that of water can be used.

30. The method as recited in claim 29, wherein said coolant material is a fluoro-polymer.

31. A method of shutting down a fuel cell system in environments subject to sub-freezing temperatures, said fuel cell system including at least one fuel cell having a membrane, a pair of gas diffusion layers, a pair of electrodes, a pair of collector plates, and at least two reactant flow lines, comprising the steps of:

reducing the fuel cell system temperature, whereby water vapor in said fuel cell is condensed;

removing water, liquid and gaseous, from said fuel cell;

purging at least one reactant gas line with a non-humidified gas; and

reducing the system pressure to a pressure approximately equal to atmospheric pressure.

32. The method as recited in claim 31, wherein the water removal step includes purging from the system.

33. The method as recited in claim 31, wherein the water removal step includes draining water to a water reservoir.

34. The method as recited in claim 31, wherein said shutdown further comprises the step of running said fuel stack in a mode that results in a pulsed current output.

35. A method of starting-up a fuel cell system in environments subject to sub-freezing temperatures, said fuel cell system including at least one fuel cell having a

membrane, a pair of gas diffusion layers, a pair of electrodes, a pair of collector plates, and at least two reactant flow lines, comprising the steps of:

flowing dry reactant gases through said at least one reactant line into said fuel cell;

measuring the temperature of said fuel cell; and

initiating humidification of said reactant gases after said fuel cell temperature is above predetermined temperature.

36. The method of claim 35, wherein said predetermined temperature is the freezing point of water at the ambient pressure.

37. The method of claim 36, further comprising the steps of :

providing a humidifier for humidifying reactant gas flows to the fuel cell stack;

providing a water reservoir for the supply of water for humidification of the reactant gas flows;

heating the reservoir to melt the water in the reservoir;

melting water in the reservoir; and

supplying water to the humidifier for humidification of the gas flows.

38. The method of claim 31 wherein the steps of heating the reservoir includes:

operating a fuel processor in an oxidant rich mode to increase heat output;

transferring a portion of said heat output to said reservoir to melt water in the reservoir;

transferring a portion of said heat output to said fuel cell to raise the fuel cell

temperature;

monitoring the melting of the water in said reservoir and the fuel cell temperature;

adjust the reactant mixture in the fuel processor to increase fuel production after at least a portion of the water in said reservoir is melted and the fuel cell temperature has reached a predetermined temperature.

39. A method of using a gasketed proton exchange membrane (PEM) fuel cell stack in sub-freezing environments, said fuel cell stack comprising a plurality of fuel cells each having a membrane, a pair of gas diffusion layers, a pair of electrodes, a pair of collector plates and at least a pair of gaskets at temperatures below the freezing temperature of water, comprising:

creating at least one flow passage through said pairs of collector plates and said pairs of gaskets of each said fuel cell not through said membrane;

flowing a coolant stream poisonous to the membrane through said at least one passage, whereby said coolant stream does not contact said membrane;

a start up including:

flowing dry reaction gases through said at least two reactant lines into said fuel cell;

measuring the temperature of said fuel stack; and

initiating humidification of said reactant gases after said fuel cell temperature is above predetermined temperature; and

a shutdown step including:

reducing the fuel cell system temperature;

purging reactant gas lines with one or more non-humidified gases; and

reducing the system pressure to a pressure approximately equal to atmospheric pressure.

40. The method of using a gasketed proton exchange membrane (PEM) fuel cell stack as recited in claim 39, said shutdown step further comprising the step of running said fuel stack in a mode that results in a pulsed current output.

41. A fuel cell stack, comprising at least one gasketed fuel cell, said at least one gasketed fuel cell comprising:

a first and a second gas diffusion layer each comprising a sheet of electrically conductive material and a pair of oppositely facing planar surfaces, wherein said gas diffusion layers are positioned approximately parallel to one another;

a membrane electrode assembly (MEA) interposed between said gas diffusion layers, said MEA having a pair of oppositely facing planar surfaces, an ion exchange membrane and a pair of electrodes interposed between said ion exchange membrane and said gas diffusion layers, said MEA extending beyond the length of said gas diffusion layers;

a pair of gaskets, wherein said MEA is interposed between the distal ends of said gaskets, said gaskets not overlapping said gas diffusion layers, wherein an interface region is produced;

said gas diffusion layers, said gaskets and said MEA all interposed within a pair

of collector plates; and,

said collector plates provide channels for reactant streams.

42. The fuel cell stack as recited in claim 41, wherein said fuel cell is a proton exchange membrane (PEM) fuel cell and said ion exchange membrane is water permeable.

43. The fuel cell stack as recited in claim 42, wherein surfaces of said channels are essentially impermeable to fluids.

44. The fuel cell stack as recited in claim 43, further comprising a pair of sub-gaskets, each sub-gasket positioned between said gaskets and extending into a position between the said gas diffusion layers and said MEA on the distal end of region where said gas diffusion layers overlaps said MEA, whereby said membrane interface region is supported.

45. The fuel cell as recited in claim 44, wherein said sub-gaskets are made materials selected from the group consisting of FEP, TFE, ETFE, PFA, CTFE, E-CTFE, PVF_2 and PVF.

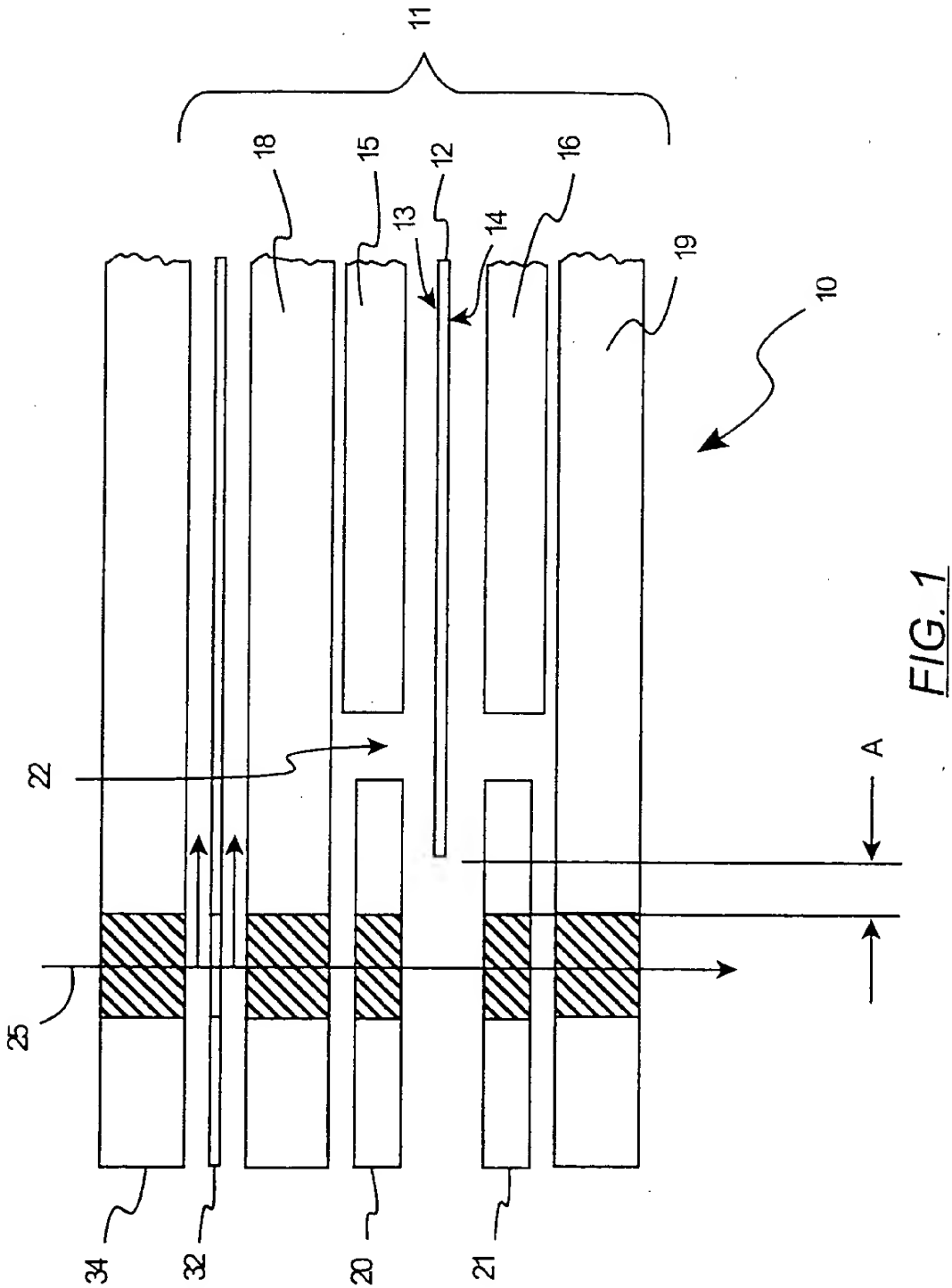

FIG. 1

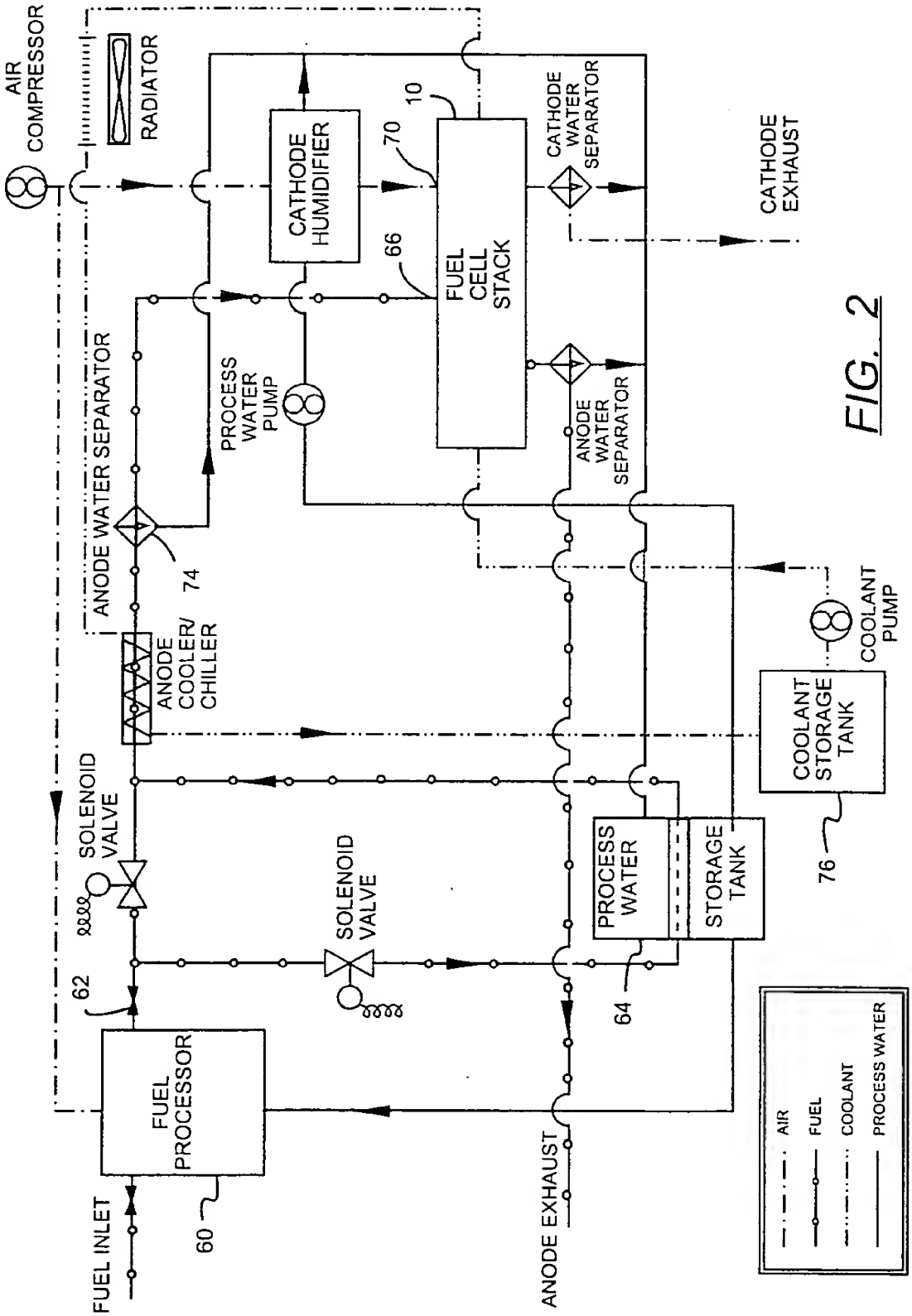
AIR COMPRESSOR
RADIATOR
CATHODE HUMIDIFIER
10
70
CATHODE WATER SEPARATOR
CATHODE EXHAUST
66
FUEL CELL STACK
ANODE WATER SEPARATOR
PROCESS WATER PUMP
ANODE WATER SEPARATOR
74
ANODE COOLER/ CHILLER
COOLANT PUMP
COOLANT STORAGE TANK
SOLENOID VALVE
SOLENOID VALVE
PROCESS WATER
STORAGE TANK
76
62
64
FUEL PROCESSOR
60
FUEL INLET
ANODE EXHAUST
AIR
FUEL
COOLANT
PROCESS WATER

FIG. 2

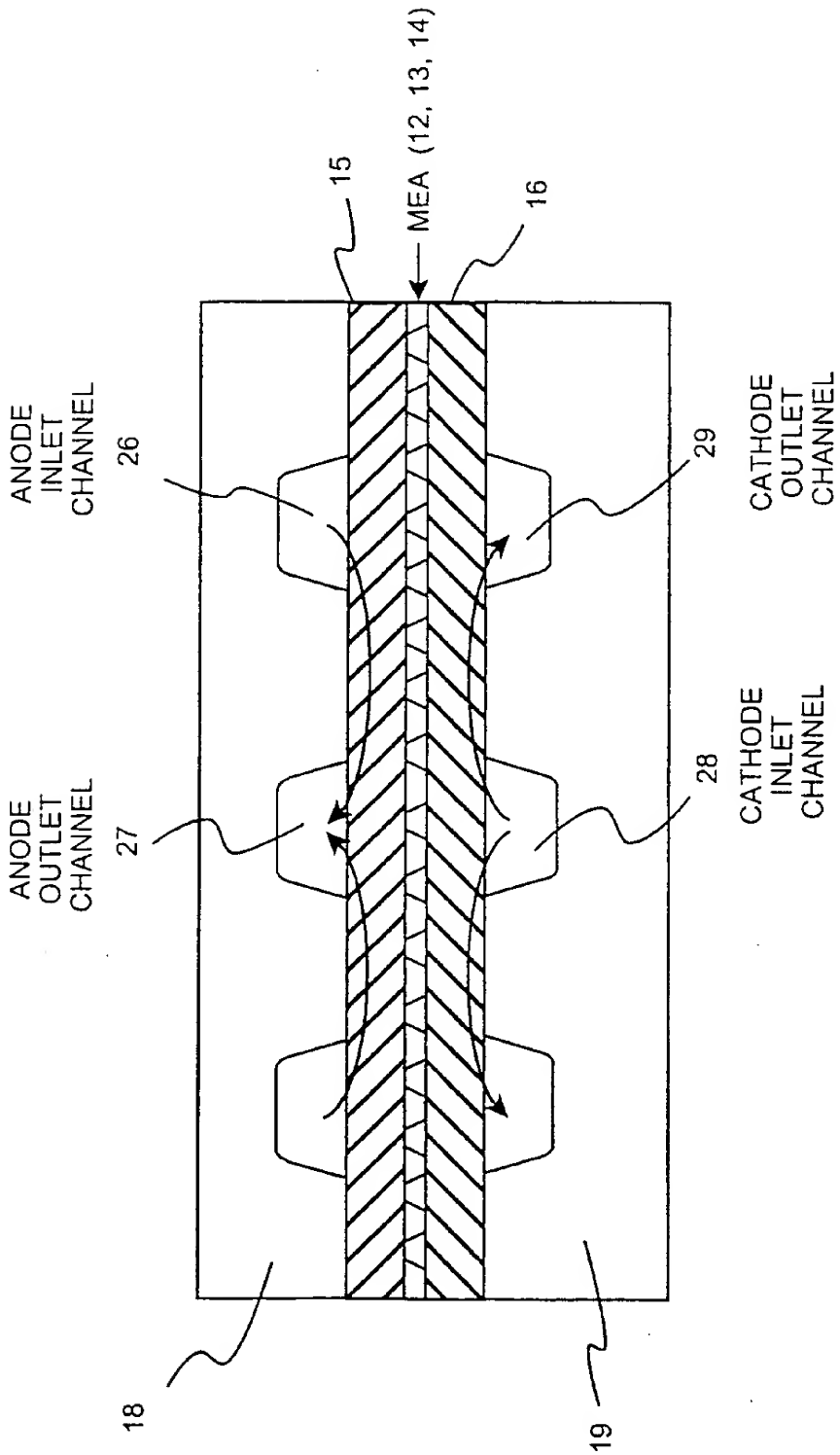
MEA (12, 13, 14)
15
16
ANODE INLET CHANNEL
26
CATHODE OUTLET CHANNEL
29
ANODE OUTLET CHANNEL
27
CATHODE INLET CHANNEL
28
18
19

FIG. 3

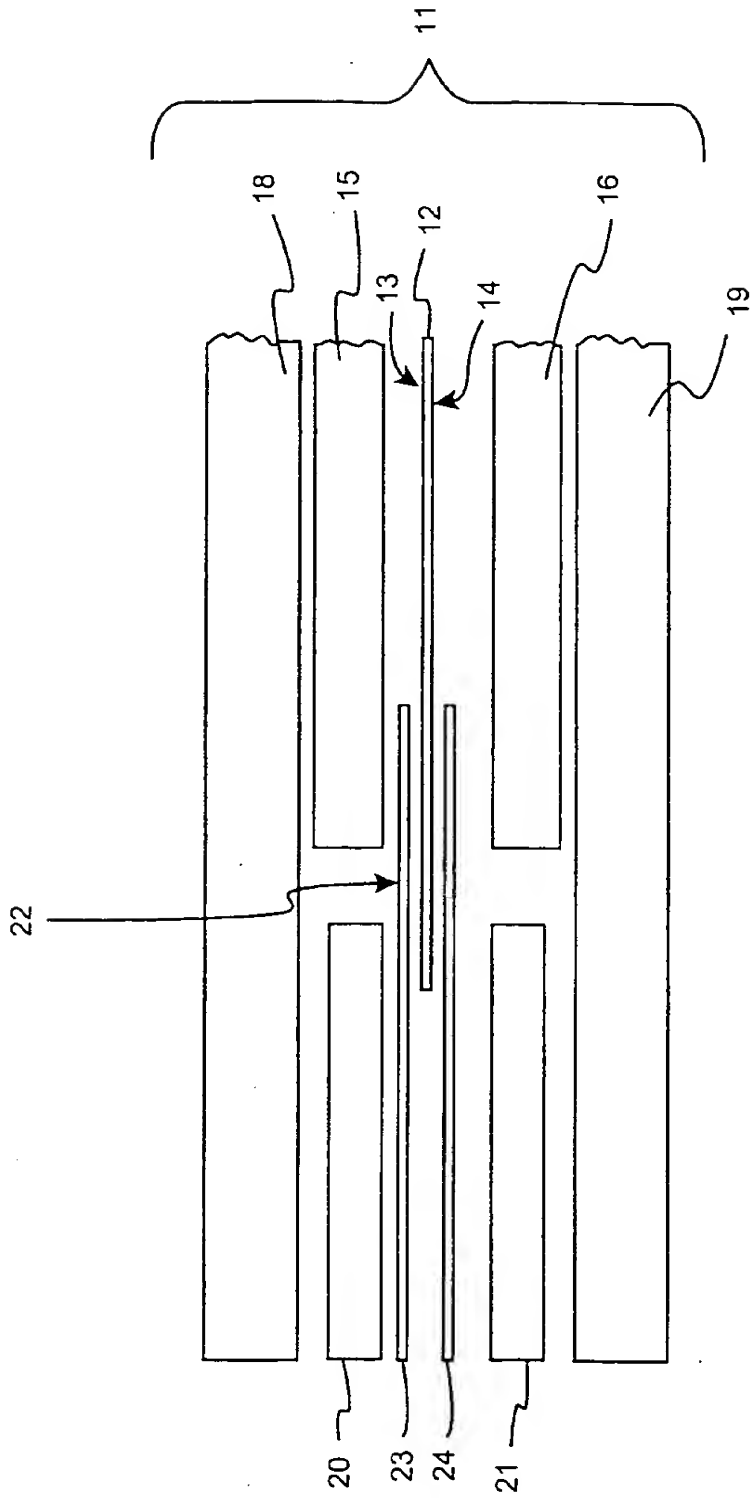

FIG. 4

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/10949

A. CLASSIFICATION OF SUBJECT MATTER
IPC(7) :H01M 8/00
US CL :429/13; 26; 34; 120
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 429/13; 26; 34; 120

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	US 5,804,326 A (CHOW et al) 08 September 1998, col. 6 line 34 to col. 7 line 5.	1, 3-13, 15-17, 22-29, 41-45 --- 2, 18-21
X --- Y	US 5,798,186 A (FLETCHER et al) 25 August 1998, col. 3 line 55 to col. 5 line 16, col. 7 lines 8-10, col. 8 line 24-35.	31-33, 35-39 --- 34 and 40

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
25 JULY 2000	30 AUG 2000

Name and mailing address of the ISA/US	Authorized officer
Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	JULIAN A. MERCADO
Facsimile No. (703) 305-3230	Telephone No. (703) 308-0661

Form PCT/ISA/210 (second sheet) (July 1998)*

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/10949

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5,863,395 A (MAH et al) 26 January 1999, column 5 lines 32-54.	14 and 30

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/10949

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

Please See Extra Sheet.

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/10949

BOX II. OBSERVATIONS WHERE UNITY OF INVENTION WAS LACKING
This ISA found multiple inventions as follows:

Group I, claim(s)1-28 and 41-45, drawn to a system.
Group II, claim(s) 29 and 30, drawn to a first method of using.
Group III, claim(s) 31-38, drawn to a second method of using.
Group IV, claim(s) 39 and 40, drawn to a third method of using.

The inventions listed as Groups I-IV do not relate to a single inventive concept under PCT Rule 13.1 because, under PCT Rule 13.2, they lack the same or corresponding special technical features for the following reasons:

Groups I-IV share the special technical feature of a fuel cell system comprising a pair of collector plates, a pair of gas diffusion layers, a membrane, a pair of electrodes, and at least one coolant passage. As this special technical feature does not define a contribution over the prior art as shown by US patent 5,804,326 to Chow et al, and no single general inventive concept exists, unity of invention is lacking.